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Recombination of hydrogen and oxygen in fluidized bed reactor with different gas distributors

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Abstract

The recombination of hydrogen and oxygen with molar ratio of 2:1 and flow rate of 2 Nm^3/h has been investigated in the fluidized bed reactor. It has been observed that the quick mixing of hydrogen and oxygen in Pt/Al_2O_3 catalyst is free of explosion and allows the effective recombination at the temperature of 373-873 K. However, this highly exothermic reaction brings the much high temperature gradient result in the fluidized bed. And the hot spot in reactor is sometimes higher than 1173K, causing the rapid deactivation of the catalyst and lead to the dangerous explosion. As adopting the multistage gas distributor, the concentration of hydrogen in the pristine gas distributor region (always in the bottom of the reactor) is effectively reduced and the temperature in the catalyst dense phase can be controlled to 673-873K, allowing a stable and effective conversion of hydrogen up to 99.9997%. The present work provides a novel fluidization technology for the recombination of hydrogen and oxygen with high concentrations.

Keywords: Recombination of hydrogen and oxygen; fluidized bed reactor; gas distributor; Pt catalyst

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1. Introduction

The recombination of hydrogen and oxygen to water is a highly exothermic reaction and an easy explosive system[1-3]. Normally, this reaction is conducted in a packed bed reactor as diluting the concentration of hydrogen beyond the explosive limits. And this way has been always adopted as treating the hydrogen gas with very low concentrations. However, in some special case, the concentration of hydrogen is very high (50-66%) and such gases are not allowed to release due to its radioactivity. The best way to treat this is to recover it to water for easy store. Apparently, such radioactivity system does not allow the introduction of inert gas to reduce the concentration of hydrogen. Thus the above requirements exert the large challenge on the catalytic conversion of such high concentration hydrogen. More seriously, the high concentration hydrogen feed gas implies the large amount of reaction heat, exerting the heavy burden on the temperature control in the reactor for safety operation. Since the heat transfer in packed bed reactor is relatively poor, it is unsafe using such reactor to convert the high concentration hydrogen[2]. In the present work, we propose a fluidized bed technology to convert the high concentration hydrogen with oxygen to water. It is observed that, as changing the type of gas distributor, we are effective to tailor the axial concentration profile of hydrogen, and consequently to control the heat release to achieve a stable temperature operation and an effective conversion of hydrogen up to 99.9997%. The results are not only useful for the treatment of special hydrogen with radioactivity, but also useful for the treatment of other hydrogen source with high concentrations.

2. Experimental

The gas source used for the experiment is pure hydrogen (99.999%) and pure oxygen (99.5%). The catalyst is a Pt/Al_2O_3 catalyst with Pt loading of 0.2%. Its average particle size is 75 micrometers and its packing density is 780 kg/m3, suitable for the gas fluidization. The reactor is a fluidized bed reactor with inner diameter of 100 mm and height of 2000 mm, made of stainless steel. The fluidized bed reactor is equipped with gas distributors, heat exchanger, and solid particle filter. Generally, there are two gas distributors in the reactor, the oxygen distributor is fixed in the bottom of reactor. And the hydrogen distributor is located on the oxygen distributor and the distance between them is 100 mm. When the oxygen is feed into the reactor, it flows up. However, the hydrogen is firstly flow upward to mix with the oxygen in the presence of the catalyst. When the temperature is controlled to 373 K, the catalyst is highly active to initialize the recombination reaction. Thus the temperature in the reactor will increase significantly. In this period, cool water is fed into the heat exchanger to control the temperature suitable for the activity range of the catalyst. With the reaction goes on, the gas is gradually converted into mostly the steam. The off gas (steam with some residue hydrogen and oxygen is first dehydrated by the cooler and the inert solid particles and finally analyzed by the online gas chromatography (TCD, HP4090D, Silica Column).

To ensure the stable and safety operation, we test two kinds of hydrogen distributor. Hydrogen distributor 1 is a simple tube distributor, which only feeds gas in an axial cross-section region of the reactor simultaneously. Hydrogen distributor 2 is a multistage tube distributor, which feeds gas in different axial cross-section region of the reactor.

Generally, the oxygen with flow rate of 1.25 Nm3/h is directly feed into the reactor. Then the flow rate of hydrogen is gradually increased from 0.35 Nm3/h to 2.5 Nm3/h. The temperature is controlled in the range of 300-1100 K. The temperature dependent conversion or the concentration dependent conversion is hydrogen is then determined. T1, T2, T3 are all the temperature spot in the catalyst dense phase, which means the region 100 ,400,700 mm above the oxygen distributor, respectively. T4 is located at the exit of the reactor, which has a inner solid particle filter nearby. Since part of catalyst particles adhere to the inner filter, it forms a special catalyst bed. Its high temperature means the insufficient conversion of hydrogen in the catalyst dense phase.

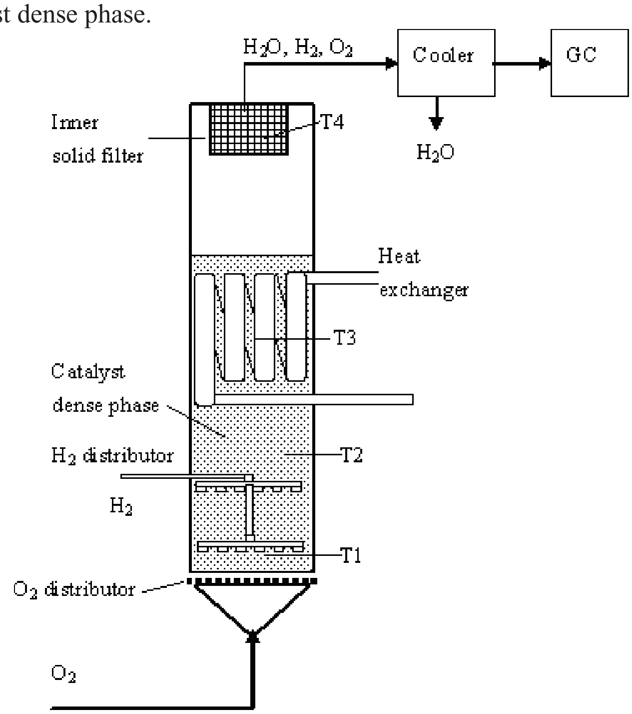

Fig.1 The fluidized bed reactor with two gas distributors

3. Result and Discussion

From Fig.2, it is clear that, when the hydrogen flow rate is smaller than 35 Nm3/h (the molar ratio of hydrogen to oxygen is 1.4:1). The temperature in the catalyst dense phase is smaller than 623 K. The process is relatively stable. However, when the flow rate of hydrogen is increased further, it is observed that the rapid temperature elevating of T1 and T2 spot to 723 K at 36 m^3/h(hydrogen) and quickly to 1120K at 42 m^3/h(hydrogen). Since the temperature of catalyst is active below 973 K. In this case, the high temperature causes the rapid sintering of Pt nano-particles on the catalyst and results in the rapid deactivation. Within several seconds, we hear the sharp explosive sound in the reactor. And the process has been stopped immediately. From this viewpoint, the reaction heat of this reaction is in large amount and the heat transfer in the reactor becomes the most important factor for the stable and effective conversion of hydrogen in this process.

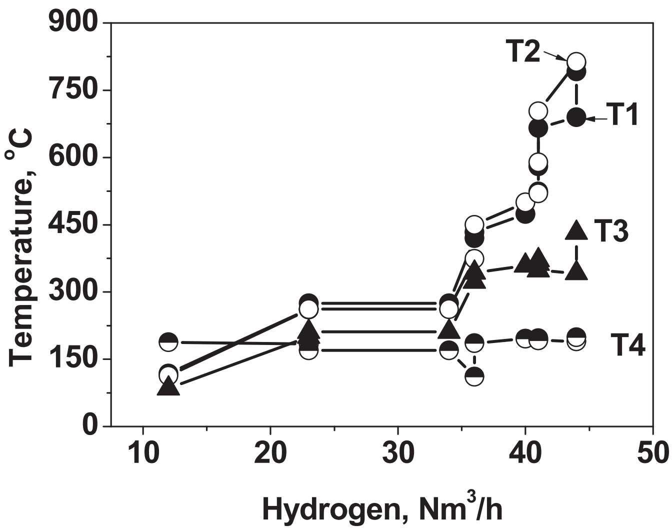

Fig.2 The axial temperature profile at different hydrogen concentrations (gas distributor 1)

As illustrated above if the hydrogen distributor 1 is used, all the hydrogen is fed into the bottom of the reactor simultaneously. Thus large amount of reaction heat is produced, and leads to the quick temperature elevating at T1 and T2 in the reactor. However, the temperature at T3 is relatively small. This means the insufficiently mixing of solid in the experimental scale fluidized bed reactor and perhaps limited heat transfer coefficient in the fluidized bed reactor. In order to overcome this bottleneck, we use hydrogen distributor 2 to fed hydrogen simultaneously at different axial cross-section of the catalyst dense phase.

It is clear that, in Fig.3, when flow rate of hydrogen is lower than 20 m3/h, we are effectively to control the temperature in the reactor lower than 6373 K. and the temperature at T1,T2,T3 is relatively close. When the flow rate and the temperature of cool water remains the same, the reaction heat will increase linearly with the flow rate of hydrogen. Thus the temperature at certain region in the reactor will also increase linearly with the flow rate of hydrogen. It is clear that, when the multistage gas distributor is adopted, the hydrogen will feed into the reactor at different region simultaneously. Thus the temperature at T1 and T2 is nearly in the same level. Since T3 is located in the upper region of the catalyst dense phase, where most hydrogen is converted, the temperature at T3 is lower than those at T1 and T3. When the gas approaches to T4, at the exit of reactor, hydrogen is nearly totally converted. Thus the temperature at T4 is nearly the same and independently of the change of flow rate of hydrogen.

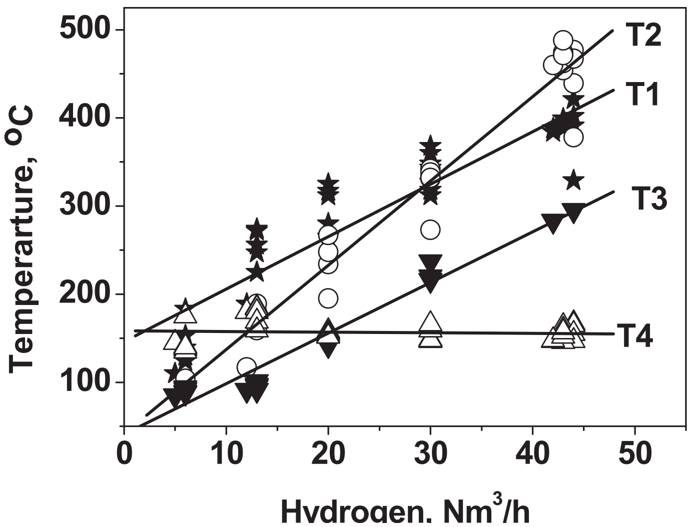

Fig.3 temperature change at different region with the hydrogen flow rate (hydrogen distributor2)

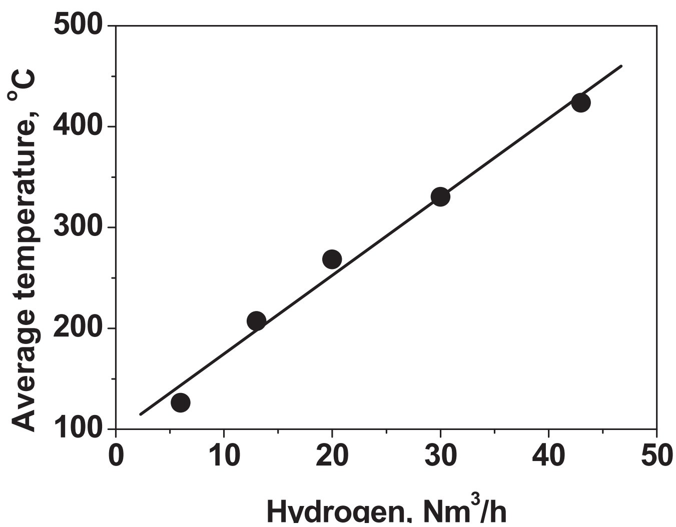

Fig.4 The average temperature at catalyst dense phase with the flow rate of hydrogen

The above analysis is in agreement with the conversion of hydrogen at different flow rate of hydrogen. It is clear that the conversion at different flow rate of hydrogen is all close to 100% (Fig.5). And our continuous operation up to 300 hours, using hydrogen distributor 2, t. indicates the process is a stable process. There is no catalyst deactivation and the fluctuation of the conversion of hydrogen. The result indicates that, adopting the novel structure of gas distributor is effective to improve the heat transfer in such large reaction heat system. Furthermore, our data indicates that the Pt/Al_2O_3 catalyst have high activity in the range of 373-873 K. If the heat transfer problem can be effectively solved, the relatively low temperature operation may be suitable for the much stable operation.

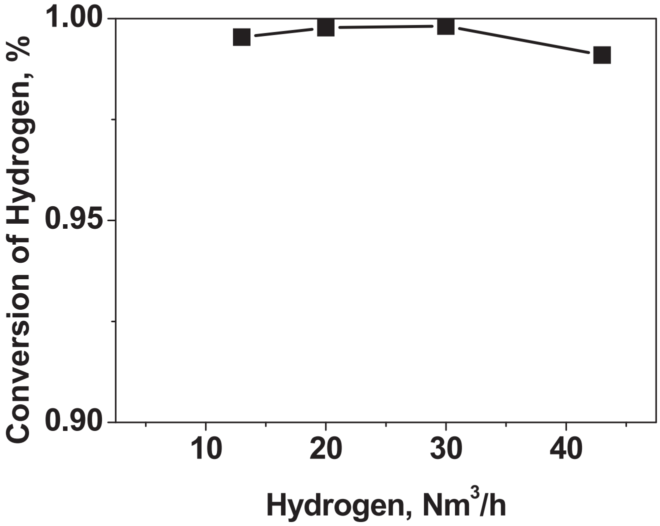

Fig.5 The conversion of hydrogen at different flow rates.

Furthermore, it is noted that our system is free of inert gas for the dilution of the hydrogen. It is an excellent catalytic example to convert such high concentration hydrogen, using a convenient way. More importantly, if the ratio of hydrogen to oxygen is 2:1, all the gas can be totally converted to water. The product water is of very small volume and easy to store. It is of significant importance when the hydrogen or oxygen is of radioactivity in certain special system.

Also notably, the product is water in the recombination of hydrogen and oxygen. And the partial pressure of water can be 100% in the system. Our results indicate that when the temperature is above the dew point of water in the system. The water does not cause serious sintering of the catalyst. However, these data are obtained at temperature below 773 K. For safety, it is necessary to study the effect of steam with high partial pressure or volume ratio on the stability of the Pt/Al_2O_3 catalyst.

4. Conclusion

The fluidized bed reactor technology is adopted to convert the hydrogen to water without addition of inert gas, using Pt/Al2O3 catalyst. As adopting a hydrogen distributor with multistage, it is effective to feed the hydrogen at different axial cross-section region of the catalyst dense phase simultaneously. Thus the temperature in such heavy exothermic reaction can be controlled beyond 773K for a long stable and effective conversion of hydrogen. The conversion of hydrogen, when the molar ratio of hydrogen to oxygen is 2:1 can be close to 100%. The fluidized bed reactor technology is effective to convert the high concentration hydrogen safely and is typically useful for such special application when the hydrogen or oxygen is of radioactivity and calls for much stable and safety operation.

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